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SUMMARY

Liquid or solid spheres levitated in film boiling on an evaporative liquid were found to follow a metastable Leidenfrost locus similar to that established for evaporative liquids "floating" on metallic surfaces. Estimates of the time a drop should remain in film boiling are much less than the recorded floating times; consequently, the drop appears to be in the metastable film boiling states. One oil/solid sphere floated on liquid nitrogen over 10 minutes. However, the maximum levitation time was not established.

On the other hand, surface protrusions and cracks on the floating spheres appeared to trigger an early termination of film boiling. Premature transition was noted for water/ice and carbon tetrachloride/solid on liquid nitrogen.

The metastable state can be terminated in one of the following heat transfer regimes: (i) transition boiling, where intermittent liquid-vapor contact caused the sphere to gyrate wildly and sink; (ii) nucleate boiling, bubbles appeared as the sphere sank; (iii) conduction and convection, where the sphere was wetted and sank. As such boule[1] behavior appears to be a limiting case of the metastable state.

A movie is available upon request.

INTRODUCTION

If a discrete amount of liquid is placed on a sufficiently hot surface, the base will evaporate so quickly that the liquid will "float" on a cushion

[1]boule: the levitation of a fluid on a sea of the same liquid prior to spontaneous coalescence.

of its own vapor. This condition is referred to as the Leidenfrost phenomenon.

According to Refs. 1 and 2 there exists a minimum surface temperature below which Leidenfrost boiling cannot be initiated. However, if film boiling is already established, the vapor cushion supporting the liquid can be maintained for surface temperatures as low as the saturation temperature, thereby completely circumventing the transition and nucleate boiling regimes of Fig. 1. This phenomenon characterizes the metastable Leidenfrost states, Ref. 3. The metastable phenomenon exists for an evaporating liquid on a heated metal surface. Can the phenomenon exist for a liquid on an evaporating liquid?

In Ref. 4, heat transfer to water/solid spheres floating on liquid nitrogen was analyzed, demonstrating the existence of the liquid on evaporative liquid Leidenfrost phenomena. However, the metastable conditions were never encountered.

Other cases of floating liquids on similar liquids are classified as boules, Ref. 5. Conditions under which boules can exist are: a physically clean surface, thermostatic control with 0.5 to 5 K superheat, a saturated vapor atmosphere, electrical neutrality, few vibrations (ref. 5). In the steady Leidenfrost state, temperature differences across the vapor cushion are large, the surface is usually contaminated, electrical charge separation can take place, and vibrations are commonplace. Boules cannot exist in this regime. However, in the metastable Leidenfrost state, where temperature differences across the vapor gap become small and vibrations and surface conditions become important, boule behavior seems possible. It appears to be a limiting case of the metastable Leidenfrost phenomenon.

The purpose of this paper is to demonstrate the existence of the metastable Leidenfrost states for liquid/solid on evaporative liquid.

THEORETICAL PREDICTIONS AND THE LEIDENFROST STATES

For a perfectly smooth surface, the vapor gap between the liquid/solid sphere and the supporting evaporating interface, see Fig. 2, theoretically is zero only when $T_s = T_{sat}$, Ref. 4. This implies that the Leidenfrost state is possible provided $T_s > T_{sat}$ and the surface remains smooth.

The floating criteria is determined by Bond number, Bo, density ratio $(\rho_s - \rho_v)/(\rho_l - \rho_v)$, and the wetting angle α, Ref. 6. In Leidenfrost boiling, the evaporative fluid does not contact the drop, which we assume to be a sphere, consequently $\alpha = \pi$. Using Fig. 3, one can determine if a given sphere will float; an approximation, within 7 percent, is provided on Fig. 3.

A simplified heat balance was used to determine the ordinary and metastable Leidenfrost time periods, see Fig. 1. The ordinary Leidenfrost period (t_L) is the sum of the times required to remove sensible heat (t_{sen}), isothermally freeze the sphere (t_f) and cool the sphere from T_f to the Leidenfrost temperature T_L, as calculated by Newton's Law of Cooling (τ)

$$\begin{aligned} t_L &= t_{sen} + t_f + \tau \\ &= R(\rho_s C_p (T_s - T_f) + \rho_{sol}\gamma)/(3\bar{h}_s(T_f - T_{sat})) \\ &\qquad - (R(\rho C_p)_{sol}/3\bar{h}_s) \ln((T_L - T_{sat})/(T_f - T_{sat})) \end{aligned} \tag{1}$$

The metastable Leidenfrost period (τ_m) is the sum of t_L and the times required to cool the sphere in temperature increments from T_L to T_{DNB}, (τ_{DNB}), from T_{DNB} to T_{incip}, (τ_{incip}), and from T_{incip} to an arbitrary temperature difference of 1° R (0.55 K). At these low ΔT's, radiation becomes important and T_s may never reach T_{sat}. In this case, equilibrium is reached and theoretically, the sphere can float indefinitely.

$$\tau_M = t_L + \tau_{DNB} + \tau_{incip} + \tau_{\Delta T = 0.55} \tag{2}$$

From Ref. 7, for nitrogen, T_L = 108 K, T_{DNB} = 94 K, and T_{incip} = 74.4 K. In these calculations, $\bar{h}_s$ is an area weighted heat transfer coefficient

$$\overline{h}_s = (1 - \cos\theta^*)h_{fb}/2 + (1 + \cos\theta^*)h_{nc}/2 - A_{rad}\, h_{rad}/A_T \tag{3}$$

where, see also Ref. 4,

$$h_{fb}\, R/k = 1 + \left[2R\overset{*}{a}\rho_s/(9(\rho_l - \rho_v)\,(\cos^2\theta^* - 2\cos\theta^* + 1))\right]^{1/4} \tag{4}$$

$$h_{nc}\, R/k \le 10 \quad \text{and} \quad A_{rad}\, h_{rad}/A_T \approx \delta(T^4_{room} - T^4_s)/(T_s - T_{sat}) \tag{5}$$

EXPFRIMENTAL PROCEDURE

The experimental apparatus is illustrated in Fig. 2. The upper and lower dishes were filled with liquid nitrogen and a drop of fluid placed on the liquid nitrogen interface of the upper dish. The floating drop was timed until it sank. The drop size was determined by comparing the frozen spheres to teflon spheres of various diameters and by direct measurement with a scale. To avoid pool nucleation sites, which cause premature transition, the dishes were carefully cleaned, and frost was minimized by use of a cover dish.

The Leidenfrost state could be terminated in any of the three regimes noted on Fig. 1; transition, or nucleate boiling, or conduction and convection. A sphere floating longer than the Leidenfrost period predicted by Eq. (1) could be sunk by ''poking'' it. If a few bubbles were noted on the surface of the sinking sphere, this would be nucleate boiling. By contrast, if vigorous bubbling and erratic surface gyrations were noted, this would be transition boiling. If the sphere would simply sink without a single bubble appearing on the surface, this would be conduction and convection.

EXPERIMENTAL RESULTS

The experimental fluids were water, carbon tetrachloride, glycerine, oil, dymethylsulfoxide, and ethylene glycol. Of these fluids, glycerine, oil, and ethylene glycol exhibited the metastable Leidenfrost phenomena, table I(a), while water, carbon tetrachloride and dymethylsulfoxide did not, table I(b).

The predicted periods were calculated from Eqs. (1) and (2), using the temperature differences appropriate to each time increment. The observed floating times of table I(a) are clearly much longer than the calculated Leidenfrost period, while those of table I(b) are not. The spheres of table I(a) appeared smooth visually, while those of table I(b) had rough surfaces. Dymethylsulfoxide exhibits metastable tendencies, however, they are quite infrequent and undependable. Metastable glycerine and oil spheres appeared to be in the glassy state, Ref. 8.

One oil/solid sphere floated over 10 minutes. However, the ultimate floating time, if such a time does exist, was not established. The floating time for a quiescent sphere stabilized in a quiescent pool could be quite long (hours) as in the case of the boule. Thus boule behavior seems to be a limiting case of the metastable states.

CONCLUSIONS

The metastable Leidenfrost phenomenon has been shown to exist for the case of liquid/solid spheres levitated on an evaporative fluid.

The metastable phenomenon can be terminated at any position along the metastable Leidenfrost line, and boule behavior appears to be a limiting case. Premature termination is caused by surface roughness. No ultimate metastable time has been established and indeed one may not exist.

SYMBOL LIST

A	Area, cm^2
Bo	Bond number = $(\rho_l - \rho_v)\, gR^2/\sigma$
C_p	specific heat, j/g-K
g	acceleration of gravity, cm/sec^2
h	heat transfer coefficient, $watt/cm^2$-K
k	thermal conductivity, j/cm-sec-K
R	sphere radius, cm

Ra*	modified Rayleigh number = $\rho_v(\rho_l - \rho_v)gR^3\ \lambda^*/(\mu k(T_s - T_{sat}))$
T	temperature, K
γ	latent heat of fusion, j/g
δ	Stefan-Boltzmann constant 5.67×10^{-12} watt/cm^2-K^4
ΔT	temperature difference, K
θ^*	submergence angle, radian
λ	latent heat of vaporization, j/g
λ^*	modified latent heat of vaporization, j/g; $\lambda^* = (1 + 0.5\ C_p(T_s - T_{sat})/\lambda)$
μ	dynamic viscosity, g/cm-sec
ρ	density, g/cm^3
σ	surface tension, dyne/cm
τ	time, (Newton's Law of Cooling), sec

SUBSCRIPTS:

f	liquid to solid transition
fb	film boiling
DNB	departure from nucleate boiling
incip	incipence of boiling
L	Leidenfrost
l	liquid (component #1)
nc	natural convection
rad	radiation
s	sphere
sat	saturation

sen	sensible (energy)
sol	solid
T	total
v	vapor (component #2)

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TABLE I. - DATA AND PREDICTED FLOATING TIMES FOR FLUID/SOLID SPHERES ON AN EVAPORATIVE FLUID (NITROGEN)

Sphere characteristics			Bond number	θ^* deg (fig. 3)	Experimental density ratio	Calculated density ratio (fig. 3)	Experimental floating time, sec	Predicted Leidenfrost period (eq. (1)), sec				Predicted metastable period (eq. (2)), sec				
Material**	Diameter, cm	Density at insertion, g/cm^3						t_{sen}	t_{ice}	τ	t_L	τ_{DNB}	τ_{incip}	$\tau_{\Delta T=0.55}$	τ_M	$\tau_{\Delta T<0.5}$
(a)																
Glycerine (glycerol)	0.38	1.25	3.18	104	1.56	1.57	250	0.35	4.2	11.9	16.5	4.2	13.9	22.1	56.7	∞
	.38		3.18	104		1.57	235	.35	4.2	11.9	16.5	4.2	13.9	22.1	56.7	Approaches
	.39		3.38	106		1.54*	340	.37	4.4	12.4	17.2	4.3	14.2	22.2	57.9	equilibrium
Oil (automotive detergent type)	0.39	0.9	3.44	90+	1.12	1.53	288	3.1	2.5	7.5	13.1	3.3	11.2	91.4	119.0	
	.39		3.44	90+		1.53	359	3.1	2.5	7.5	13.1	3.3	11.2	91.4	119.0	
	.39		3.38	90+		1.54	619	3.	2.5	7.5	13.0	3.3	11.1	87.2	111.0	
Ethylene-glycol	0.5	1.113	2.0	93	1.39	1.88	177	1.7	3.2	7.4	12.3	3.0	10.1	18.4	43.9	
	.3		2.0	93	1.39	1.88	472	1.7	3.2	7.4	12.3	3.0	10.1	18.4	43.9	
	.35		2.72	94		1.86	146	1.7	3.2	7.4	12.3	3.0	10.0	17.0	42.5	
(b)																
Water	0.6	1	8.0	110	1.25	1.3	Average 23 to 28 sec	3.9	12.4	16.3	32.6					
	.56		7.0	104		1.32		3.6	11.4	14.9	29.9					
	.52		5.6	98.9		1.41		3.0	9.8	12.8	25.6					
Carbon tetrachroride	0.22	1.59	1.07	94.5	1.99	2.7	Average 0 to 10	.7	.3	1.8	2.8					
	.2		1.08	93.0		2.7		.6	.2	1.6	2.4					
	.18		.72	92.0		3.45		.5	.2	1.3	.2.0					
Dymethyl-sulfoxide $(CH_3)_2SO$	0.32	1.1	2.3	93.4	1.37	1.9	Average 8-14	0	1.5	1.0	2.5					
	.3		2.0	92.6		2.0		0	1.3	.9	2.2					
	.28		1.7	92.0		2.13	44+	0	1.2	.8	2.0					
Blood	0.41	~1	3.7	94.0	1.25	1.6	16	2.3	7.3	9.6	19.2					
	.28		1.73	92.0		2.1	23 to 30+	1.3	4.1	5.4	10.8					
	.2		.9	92.0		3.	23 to 26+	.8	2.4	3.2	6.4					
	.16		.57	91.		4.	21	.5	1.7	2.3	4.5					

All spheres of table I(b) terminated in nucleate or transition boiling-except the $(CH_3)_2SO$, 44 sec-sphere which appeared to be metastable

*Use figure 3

**Solid properties ill defined

+Did not roll on the surface and few, if any, bubbles appeared as it sank. Crystal structure and entrapped air greatly alter floating character

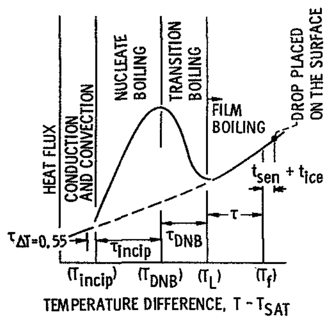

Figure 1. - Conventional boiling curve with metastable Leidenfrost states (– – –).

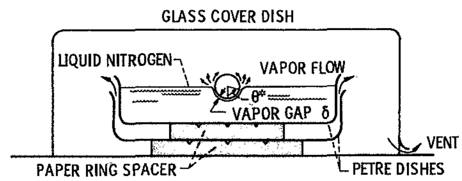

Figure 2. - Schematic of the experimental apparatus and model of a sphere supported by an evaporative fluid interface.

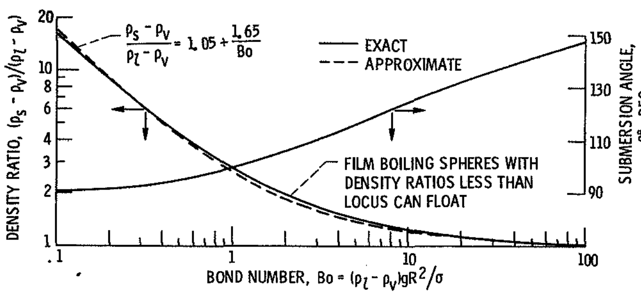

Figure 3. - Optimum levitation Loci for spheres in Leidenfrost film boiling. Maximum density ratio and submergence angle variation with Bond number.